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(54) Title: MULTILAYER POLYMERIC BODY WITH INTERFERENCE EFFECT

(57) Abstract

The invention relates to a multilayer reflective polymeric body of at least first and second diverse generally transparent polymeric materials, arranged in a sufficient member of alternating layers of said first and second polymeric materials such that a definite part of the light incident on said body is reflected, wherein said first and second polymeric materials differ from each other in refractive index by at least about 0.03 for the screening of solar radiation with the aim of influencing plant growth and the morphogenesis.

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Multilayer polymeric body with interference effect

The present invention relates to a multilayer polymeric film with interference effect. More particularly, the present invention relates to an agricultural film for the screening of solar radiation with the aim of influencing plant growth and the morphogenesis.

It is necessary for a controllable growth of plants to set up special light conditions to affect plant growth and morphogenesis. Special light conditions for plant growth, for example, within a greenhouse or under a mulch film include some which are of particular interest. The temperature within the enclosed area as well as the light intensity and the spectral distribution of the incoming light are important factors for the plant growth.

Plants in a greenhouse, for example, will not grow so well if the temperature variation between day and night is large due to a big difference in visible light and NIR radiation impact.

From this it follows that the plants stay back in growth. Thus, for the photosynthesis in greenhouses the photosynthetically active radiation (PAR) is only desired while other parts of the solar light should be screened because they have mainly a negative influence on the microclimate. Therefore the following objectives have to be achieved: Firstly, the intensive irradiation by sunlight in particular by NIR radiation needs to be screened, in order to avoid too high temperatures within the enclosed area. Secondly, the incoming light beams are to be scattered in order to prevent burnings on plants. Thirdly, heat within the area is to be stored at night.

However, in case of mulch films the opposite effect is desired. The PAR should be screened and all other parts of the solar light should be transmitted. Mulch films are used in agriculture to improve the growing conditions of the useful plants and to minimize the use of chemical herbicides.

For certain crop growers want to intervene in the plant morphogenesis. Morphogenesis is the influence of environmental factors on the shape and appearance of the plant. One of these factors is the spectral energy distribution.

Raviv and Allingham describe in "Plastic Culture", No. 59, September 1989, pp. 3 to 12, that diffuse light is favourable for a steady growth of plants. Furthermore, it turned out that diffuse light has the advantage of a more uniform illumination and that it does not damage the plants as much as direct radiation. Hancock describes in "Plastic Culture", No. 79, 1988, pp. 4 to 14, that special additives in LDPE have a favourable effect on the microclimate in greenhouses because of the generation of diffuse light and the creation of a favourably thermic effect in the greenhouses. Suitable additives are aluminosilicate in the form of kaolin, calcium carbonate, talc and kaolin clay.

Greenhouses made of glass already fulfil some of the above-mentioned conditions by virtue of the inherent physical properties of inorganic glasses. Furthermore, constructive measures, for example knurled glass or shadow cloth are known.

If the enclosing material is plastic, such as, for example, polyethylene, polyethylene/vinyl acetate copolymers, polyvinyl chloride, polycarbonate or polyacrylate, it is much more difficult to meet these requirements. The high transparency of many plastics for IR radiation results in good heat transmission which, in the case of films, is additionally assisted by the low film thickness.

A polymethyl methacrylate glazing material for buildings and vehicles containing an interference pigment for the screening of NIR radiation of a wave length between 800 and 1500 nm is disclosed in DE 2,544, 245.

The pigment used has a blue-red colour and the light transmitted through the glazing material has a yellow-green colour. When used in greenhouses, this glazing material has the disadvantage that portions of visible light which cannot be utilized by the plant are transmitted and, on the other hand, the red portion of visible light which can be utilized by the plant is screened off by said glazing material. Furthermore, the transmitted green portion has the additional disadvantage that it contributes to heating of the greenhouse by virtue of its conversion into long wave light.

EP-A 0,428,937 describes a grey-white coating composition for greenhouses consisting of a polymer substrate and reflecting particles suspended therein. These particles are aluminium platelets or mica platelets coated with titanium dioxide. The coating composition is used for temporary coatings in extreme weather conditions (summer). In winter, it can again be removed by washing it off with a water jet. More details regarding the pigment used are not given.

The coating composition has the disadvantage that not only the IR beams but also a substantial portion of the visible light which can be utilized by the plant are reflected by the metal particles. The green portion of the transmitted light cannot be utilized by the plant and contributes to heating of the greenhouse.

EP-A-03 98 243 describes polymeric mulch sheets and mulch films for use in agriculture which have a green absorption colour and contain a UV stabilizer. The green sheets and films absorb a large part of the solar radiation which promotes photosynthesis and plant development (PAR), and transmits enough solar radiation so as to heat the soil beneath such films and sheets. Such mulch films and sheets reduce weed growth but they have the disadvantage that the photosynthetically active radiation is absorbed and is lost for the useful plant.

L.M. Mortensen and E. Stromme describe in Scientia Horticulturae 33 (1987), 27-36 the effects of light quality on some greenhouse crops. Different light qualities were established in growth chambers placed outdoors by selective screening of the natural light spectrum in the chambers. The PAR level was the same at all light qualities. Blue light containing a high red/far-red ratio reduced the dry weight compared to natural (N), green, yellow and red light in chrysanthemum, tomato and lettuce. Plant height in chrysanthemum and tomato was strongly reduced by blue compared to N light, and strongly increased by green and yellow light.

WO 94/05727 discloses a composite material for the selective screening of radiation comprising a polymer and an interference pigment. The composite material can be employed in form of films, sheets and webs as covering for greenhouses, for the influencing of plant morphogenesis and for mulching purposes.

Surprisingly, it has been found that a multilayer reflective polymeric body of at least first and second diverse generally transparent polymeric materials, arranged in a sufficient member of alternating layers of said first and second polymeric materials such that a definite part of the light incident on said body is reflected wherein said first and second polymeric materials differ from each other in refractive index by at least about 0.03 can be employed for the screening of solar radiation with the aim of influencing plant growth and the morphogenesis.

The polymeric body can be used in the form of a film, sheet or web.

Multilayer reflective polymeric bodies and their manufacture are well known and described in US patents Nos. 2 408 398, 2 790 202, 2 820 249, 2 902 754, 3 024 494, 3 147 515, 3 308 508, 5 217 794 and 5 234 729 and in Polymer Engineering and Science 1969, Vol. 9, No. 6, pages 400-404 under the title "Physical Optics of Iridescent Multilayered Plastic Films" by T. Alfrey, E.F. Gurnee and W.J. Schrenk, in a brochure of The Dow Chemical Company titled "Coextrusion of Blown Multilayer Plastic Films" by ".J. Schrenk and T. Alfrey, and in Journal of Plastic Film & Sheeting 1994, Vol. 10, pages 78-79 by J.A. Wheatley and W.J. Schrenk.

Normally, said polymeric body has an iridescent or silvery appearance but by keeping definite tolerances of the layer thickness of about 5 nm it is possible to reflect a definite part of the light incident on the body. In this case the polymeric body appears in a definite color in the reflected light and in the complementary color in the transmitted light.

By variation of layer thickness, number of the alternating layers and the polymeric material the desired color can be adjusted. That means the reflecting band can intensionally be shifted in the spectrum of the sunlight.

The polymeric material used for the preparation of the composite materials are transparent materials like polyolefins, co- or terpolymers, for example low-density polyethylene (LDPE), vinylester copolymers like ethylene-vinylacetate copolymer (EVA), fluoropolymers, co- or terpolymers and polyvinylidene chloride (PVDC), polyvinyl chloride (PVC), polycarbonate (PC), polymethylacrylate (PMMA), polystyrene (PS), polyesters (PET, PBTE) or mixtures of transparent polymers. The term "polymer" may include mixtures of polymers.

The polymeric material has to be choosen so that the difference in refractive index is bigger than .03.

The term multilayer reflective polymeric body includes noncontinuous layers of films, such as lamellar or particulate structures incorporated into a polymer matrix.

The protective skin layer improves the mechanical, optical and application properties such as weather resistance, UV stability, and non-drop behavior.

The number of alternating layers of the polymeric body can be varied between 30 and 3000, preferably between 50 and 1000. The total number of layers for the desired effect depends on the difference of refractive indices. The smaller the difference the higher the total number of layers.

The entire disclosure of all applications, patents and publications, cited above and below are hereby incorporated by reference.

In order that the invention may be more readily understood, reference is made to the following example, which is intended to be illustrative of the invention, but is not intended to be limiting in scope.

Example 1

Fig. 1 shows the reflectance spectrum of a polymeric multilayer film which consists of alternating layers of polycarbonate (PC refractive index n_D = 1,587) and polymethacrylate (PMMA, n_D = 1,491). The total numbers of layers is 100 and the layer thickness is 100 nm for a PC layer resp. 140 nm for PMMA layer.

The spectrum shows that the film is transparent below 700 nm and reflects with a maximum around 740 nm.

This film therefore is suitable for influencing of the growth of plants, such as vegetables and tomatoes, so that this plants become more compact. The background for this photomorphogenetic effect is the change of the red to far red ratio for the phytochrome.

Example 2

Fig. 2 shows the refractive spectrum of a polymeric multilayer film which consists of alternating layers of PC and PMMA. The total number of layers is 100 and the layer thickness is 100 nm for a PC layer resp. 115 nm for PMMA layer.

This film is transparent below 620 nm and above 700 nm and shows reflection inbetween. This film is suitable for influencing the growth of plants, when slender plants are desired, such as for roses.

The background here is the opposite effect of Example 1.

Example 3

Fig. 3 shows the reflective spectrum of a polymeric multilayer film which consists of alternating layers of PC and PMMA. The total numbers of layers is 50 and the layer thickness is 70 nm for PC resp. 105 nm for PMMA. This film reflects the green light around 540 nm that cannot be utilized for the growth of plants (minimum of PAR). It is therefore suitable for greenhouse films.

Example 4

Fig. 4 shows the reflective spectrum of a polymeric multilayer film which consists of alternating layers of PC and PMMA. The total number of layers is 500 and the layer thickness is (70 + 0.05 n) nm for PC and (75 + 0.0533 n) nm for PMMA, n indicating the number of layer.

This spectrum is similar to the reflection spectrum in T. Alfrey, jr. et al. Polym. Eng. Sci. 9 (1969), 400-404. This film reflects nearly the total PAR spectrum and therefore suitable for mulch foils.

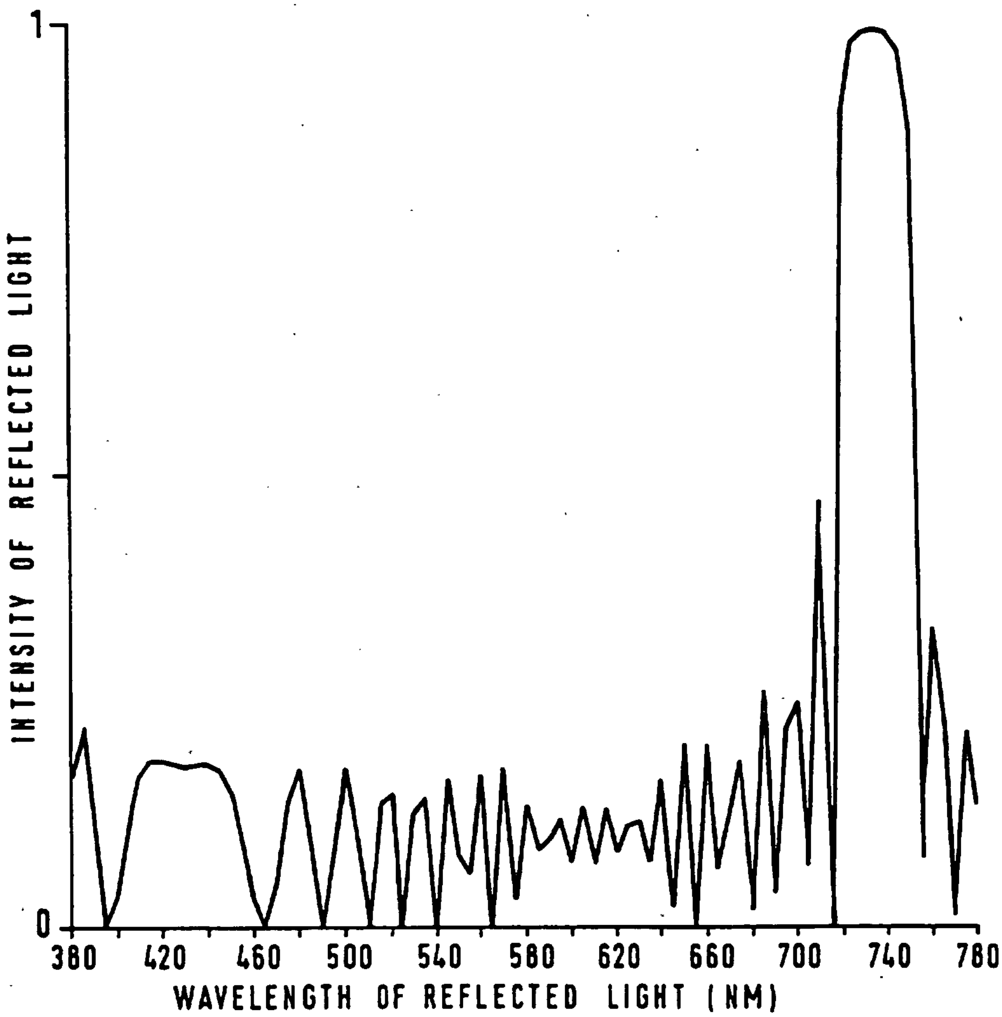
Claims

1. A multilayer reflective polymeric body of at least first and second diverse generally transparent polymeric materials, arranged in a sufficient member of alternating layers of said first and second polymeric materials such that a definite part of the light incident on said body is reflected, wherein said first and second polymeric materials differ from each other in refractive index by at least about 0.03 for the screening of solar radiation with the aim of influencing plant growth and the morphogenesis.

2. The multilayer reflective polymeric body of claim 1 wherein said polymeric body is in the form of a film, sheet or web.

3. The multilayer reflective polymeric body of claim 1 wherein said polymeric body includes a protective skin layer on at least one major surface thereof.

4. The multilayer reflective polymeric body of claims 1 to 3 wherein said protective skin layer contains pigments, dyes, fillers, antidrop agents and functional additives and processing aids necessary for the particular polymer.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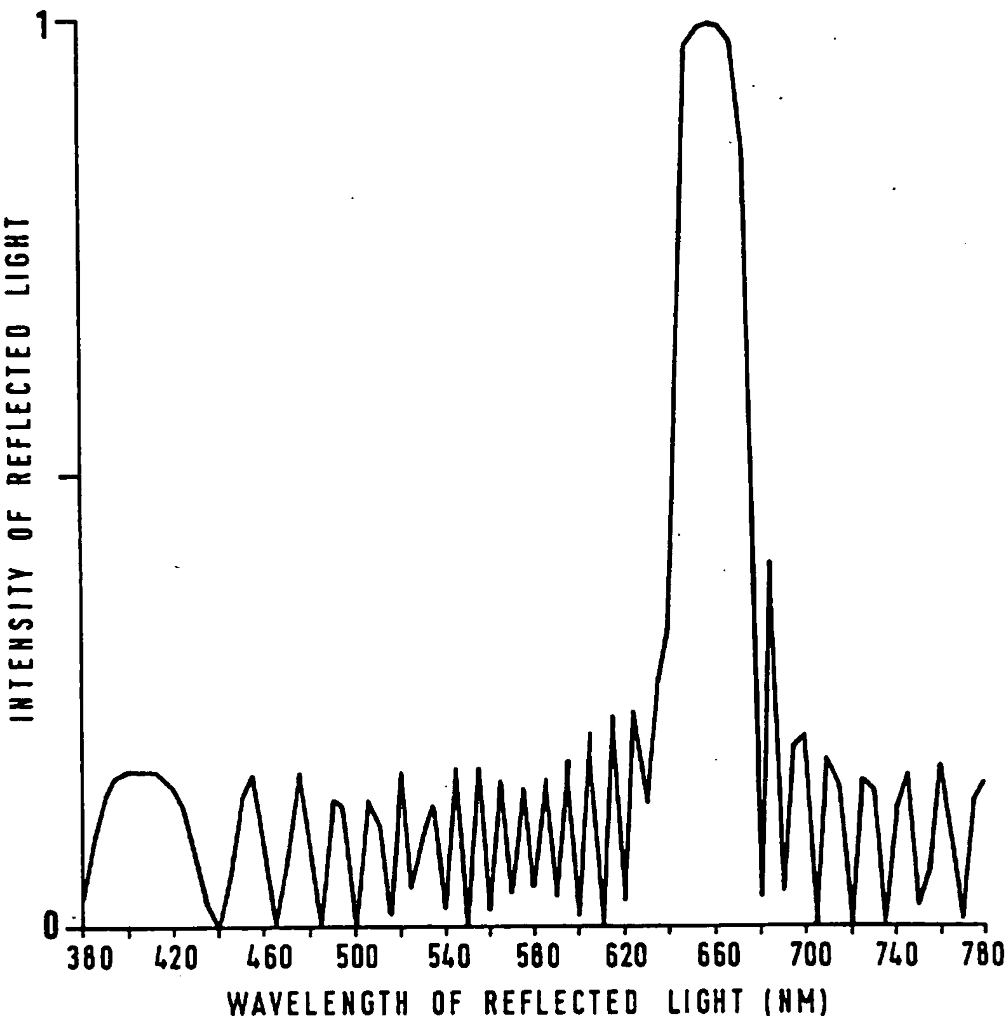
INTENSITY OF REFLECTED LIGHT
380 420 460 500 540 580 620 660 700 740 780
WAVELENGTH OF REFLECTED LIGHT (NM)

FIG. 1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INTENSITY OF REFLECTED LIGHT
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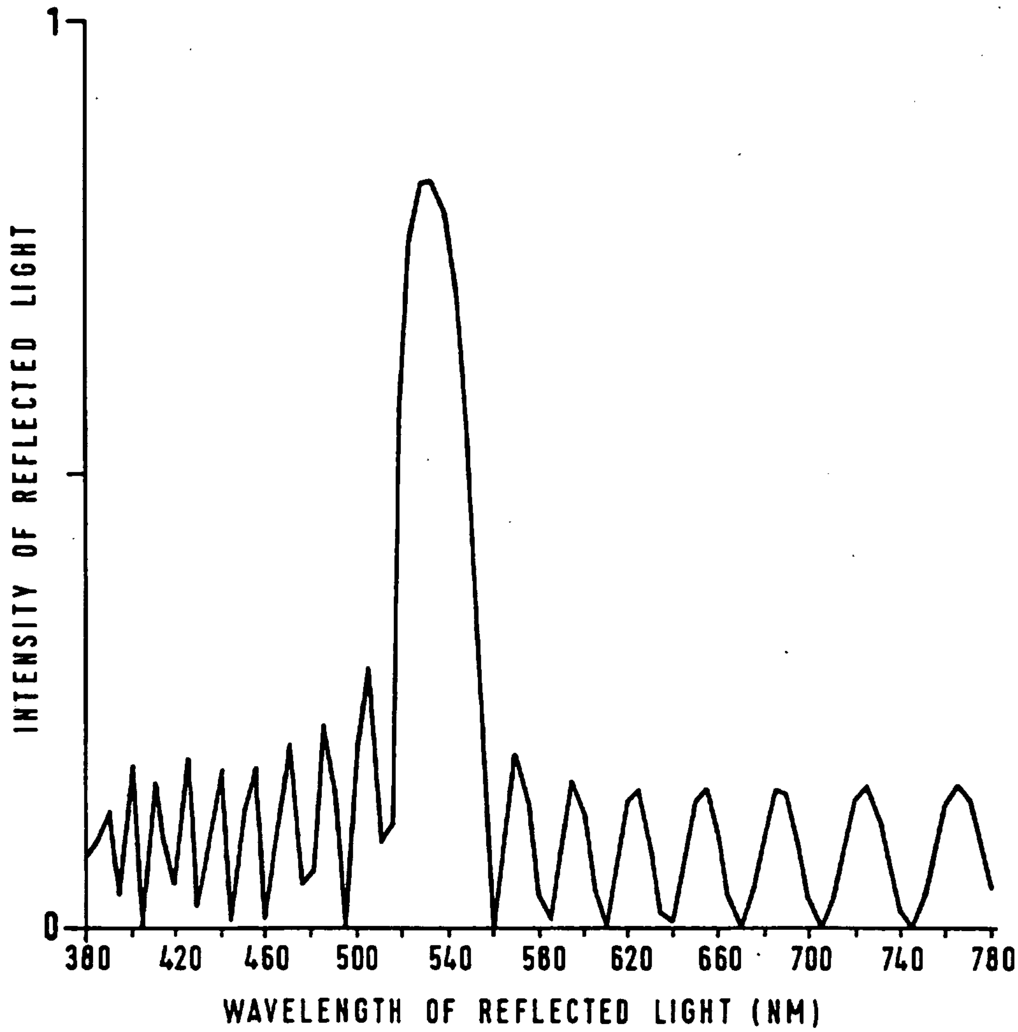
420
460
500
540
580
620
660
700
740
780
WAVELENGTH OF REFLECTED LIGHT (NM)

FIG. 2



1
0
INTENSITY OF REFLECTED LIGHT
380
420
460
500
540
580
620
660
700
740
780
WAVELENGTH OF REFLECTED LIGHT (NM)

FIG. 3

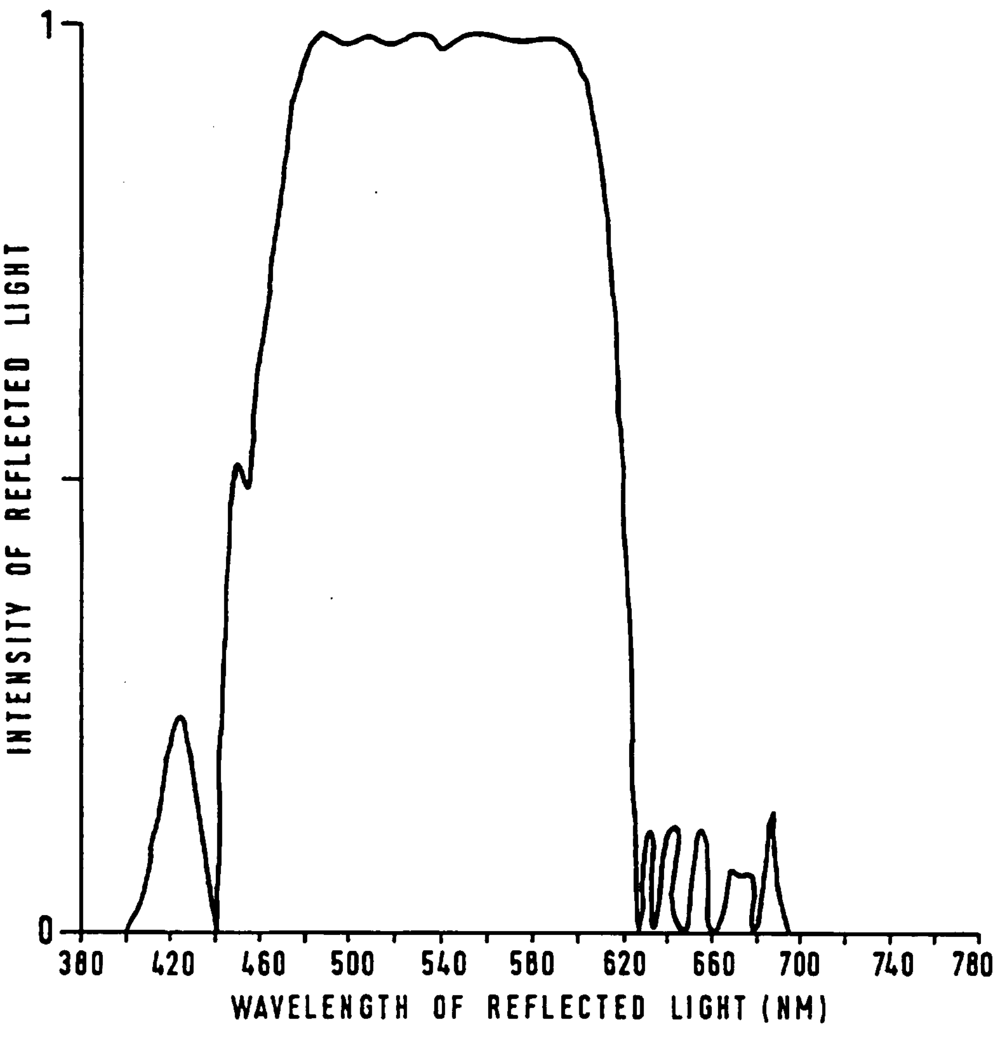

1
0
INTENSITY OF REFLECTED LIGHT
380 420 460 500 540 580 620 660 700 740 780
WAVELENGTH OF REFLECTED LIGHT (NM)

FIG. 4

INTERNATIONAL SEARCH REPORT

International Application No PCT/EP 95/02786

A. CLASSIFICATION OF SUBJECT MATTER
A 01 G 13/02

According to International Patent Classification (IPC) or to both national classification and IPC 6

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
A 01 G,B 32 B,C 08 J,C 08 K,C 09 D,G 02 B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP, A, 0 298 942 (TENO AB) 11 January 1989 (11.01.89), claims 5,8,9; fig. 4; abstract. --	1-4
A	EP, A, 0 455 942 (CARL FREUDENBERG) 13 November 1991 (13.11.91), abstract. --	1,2,4
A	WO, A, 86/07 235 (GRAVISSE) 18 December 1986 (18.12.86), abstract. --	1,2
A	US, A, 4 920 692 (KITAMURA et al.) 01 May 1990 (01.05.90),	1-4

[X] Further documents are listed in the continuation of box C.

[ ] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
17 October 1995	23.11.95

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INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP 95/02786

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	abstract; claims.	
A	EP, A, 0 398 243 (GINEGAR PLASTIC PRODUCTS) 22 November 1990 (22.11.90), the whole document (cited in the application).	1-4
A	WO, A, 94/05 727 (HYPLAST N.V.) 17 March 1994 (17.03.94), abstract; claims (cited in the application).	1-4
A	EP, A, 0 428 937 (STATE OF ISRAEL MINISTRY OF DEFENCE) 29 May 1991 (29.05.91), abstract; claims (cited in the application).	1,4
A	DE, A, 2 544 245 (RÖHM GMBH) 14 April 1977 (14.04.77), the whole document (cited in the application).	1-4
A	US, A, 3 147 515 (DONALD L. AMSDEN) 08 September 1964 (08.09.64), the whole document (cited in the application).	1

ANHANG

zum internationalen Recherchenbericht über die internationale Patentanmeldung Nr.

ANNEX

to the International Search Report to the International Patent Application No.

ANNEXE

au rapport de recherche international relatif à la demande de brevet international n°

PCT/EP 95/02786 SAE 114401

In diesem Anhang sind die Mitglieder der Patentfamilien der im obengenannten internationalen Recherchenbericht angeführten Patentdokumente angegeben. Diese Angaben dienen nur zur Unterrichtung und erfolgen ohne Gewähr.

This Annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The Office is in no way liable for these particulars which are given merely for the purpose of information.

La presénte annexe indique les membres de la famille de brevets relatifs aux documents de brevets cités dans le rapport de recherche international visée ci-dessus. Les reseignements fournis sont donnés à titre indicatif et n'engagent pas la responsibilité de l'Office.

Im Recherchenbericht angeführtes Patentdokument / Patent document cited in search report / Document de brevet cité dans le rapport de recherche	Datum der Veröffentlichung / Publication date / Date de publication	Mitglied(er) der Patentfamilie / Patent family member(s) / Membre(s) de la famille de brevets	Datum der Veröffentlichung / Publication date / Date de publication
EP A1 298942	11-01-89	SE A0 8702819	09-07-87
		SE A 8702819	10-01-89
EP A2 455942	13-11-91	AT E 110223	15-09-94
		DE A1 4014717	14-11-91
		DE C2 4014717	19-11-92
		DE C0 59102606	29-09-94
		EP A3 455942	22-04-92
		EP B1 455942	24-08-94
		ES T3 2058949	01-11-94
WO A1 8607235	18-12-86	DE C0 3678800	23-05-91
		EP A1 224534	10-06-87
		EP B1 224534	17-04-91
		FR A1 2582471	05-12-86
		FR B1 2582471	29-06-90
		JP T2 63500213	28-01-88
US A 4920692	01-05-90	JP A2 62265927	18-11-87
		JP B4 4058934	18-09-92
		AU A1 68288/87	11-02-88
		AU B2 596637	10-05-90
		CN A 87100858	17-02-88
		CN B 1009793	03-10-90
		DE C0 3767402	21-02-91
		EP A1 256201	24-02-88
		EP B1 256201	16-01-91
		KR B1 9500683	27-01-95
		SG A 363/91	21-06-91
EP A1 398243	22-11-90	IL A0 90321	15-12-89
		US A 5138792	18-08-92
WO A2 9405727	17-03-94	CN A 1083829	16-03-94
		DE A1 4230023	17-03-94
		EP A1 659198	28-06-95
		FI A 951074	08-03-95
		FI A0 951074	08-03-95
		IL A0 106759	08-12-93
		WO A3 9405727	09-06-94
		NL A 9201568	05-04-94
EP A1 428937	29-05-91	AU A1 66692/90	23-05-91
		CA AA 2029941	21-05-91
		IL A0 92372	26-07-90
		ZA A 9009177	25-09-91
DE A1 2544245	14-04-77	AT A 5157/76	15-04-80
		AT B 359720	25-11-80
		CA A1 1085205	09-09-80
		DE B2 2544245	30-04-81
		DE C3 2544245	17-12-81
		FR A1 2326549	29-04-77
		FR B1 2326549	25-06-82
		IT A 1069666	25-03-85
		NL A 7610912	05-04-77
		US A 4090773	23-05-78
US A 3147515		keine - none - rien	